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Integrated Performance of Fenton Process and Filtration (Activated Charcoal and Sand) for Textile Wastewater Treatment

Md. Sabbir Hossain¹, Protima Sarker^{2*}, Md. Shiblur Rahaman² and Md. Khabir Uddin³

¹Department of Environmental Science and Technology, Jashore University of Science and Technology, Jashore-7408, Bangladesh. ²Department of Environmental Science and Disaster Management, Noakhali Science and Technology University, Noakhali-3814, Bangladesh. ³Department of Environmental Sciences, Jahangirnagar University, Dhaka-1342, Bangladesh.

Authors' contributions

This work was carried out in collaboration among all authors. Author MSH designed the study, performed the statistical analysis, managed the analyses of the study. Author PS managed the literature searches, wrote the protocol and wrote the first draft of the manuscript. Author MSR reviewed and revised the manuscript. Author MKU supervised the study. All authors read and approved the final manuscript.

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ABSTRACT

Textile effluents contain complex characteristics with different physicochemical parameters and colorful dye. It is difficult to remove all types of pollutant using a specific treatment process. In this study adsorption filtration alone then Fenton process with sand-gravel mixed filtration and activated charcoal filtration was applied. Fenton process is very effective process for Chemical Oxygen Demand (COD) and Dissolve Organic Carbon (DOC) removal. And Activated carbon is a suitable adsorbent for dye removal. Physicochemical parameters like, COD, DOC, Total Dissolve Solid (TDS), Dissolve Oxygen (DO), Electric conductivity (EC), and pH was measured before and after the treatment process. After filtration by adsorbent filter all the parameter reduced than raw effluent

^{*}Corresponding author: E-mail: protima_das37@yahoo.com;

but when Fenton process with sand filtration and activated charcoal filtration was applied all the parameter drastically changed than before. DO level was increased after only adsorption filtration process, Fenton and normal sand filtration process, Fenton, and adsorbent filtration process as 1.8 mg/l < 5 mg/l < 6.19 mg/l < 6 mg/l, respectively. Maximum level of EC (61.33%), TDS (82.75%), BOD (92%), COD (80%), DOC (97.57%), and color (78.57%) was decrease after using Fenton process with Activated carbon filtration process than other treatment process, which was within Bangladesh standard value. This result represents the advantage of using Fenton process with adsorption filtration to treat textile effluent. Moreover, this process is easily operating, practical and environment friendly for using in industrial scale.

Keywords: Advance oxidation process; adsorption; chemical oxygen demand; dissolve organic carbon; textile effluent.

1. INTRODUCTION

Textile industries are considered as the largest industrial sectors of Bangladesh as 78% of the total export earning comes from textile and textile related goods contributing 12% to GDP [1]. But industrial wastewater imposes serious impact on aquatic ecosystem and human health. Because it generates high concentration of polluted water discharge into water body and without inadequate treatment process Major [2]. pollutants in textile wastewaters are high suspended solids, oxygen consuming matter, heat, color, acidity and other soluble substances [3]. Highly colored wastes prevent light penetration and may disturb the ecosystem. Moreover, dyes itself are toxic to some organism [4].

According to Environmental Conservation Act (1995) and Environmental Conservation Rules (1997) [5], Bangladesh, it is necessary to treat wastewater prior to discharge in water body to prevent the adverse effect regarding to textile wastewater. From previous research it has been confirmed that physico-chemical processes (e. coagulation/flocculation. precipitation, g., adsorption, ion exchange, membrane separation and oxidation) is not effective for complete remediation [6] and biological methods are inefficient especially if the pollutants are present concentrations Recently, in high [7]. electrochemical technologies and Advanced Oxidation Processes (AOPs) (such as Fenton, Photo- Fenton process) have been considered as efficient alternatives for textile wastewater Advanced treatment [8]. [9]. oxidation processes (AOPs) show promising results in polluted highly treating wastewater [10]. Fenton process is one of the advanced oxidation processes which yields HO when reacting with iron (Fe²⁻), which acts as a catalyst and a strong oxidant such as hydrogen peroxide

 (H_2O_2) and is a frequently investigated oxidation process [11].

$$Fe^{2^+}+6H_2O \rightarrow [Fe(H_2O)_6]^{2^+}$$
 (1)

$$[Fe(H_2O)_6]^{2^+}+HO^- \rightarrow [Fe(H_2O)_5(OH)]^++H_2O$$
 (2)

$$[Fe [(H_2O)_5 (OH)]^+ + hv \to Fe^{2+} + 5H^+ + 6HO^{\bullet} (3)$$

The Fenton oxidation is an attractive method to mineralize dyes remaining in textile wastewater. Indeed, H_2O_2 is an environmentally friendly oxidant, which is decomposed into water and oxygen, and iron is highly abundant and presents low toxicity [7]. Moreover, this process is a low cost, time saving and effective technology for textile effluent treatment [12]. Fenton oxidation has been successful in degrading most of the organic contaminants [13],[14],[15]. But for dye wastewater which presented as mixtures with different physical and chemical characteristics, there are still some limitation factors, such as high suspended solids, excessive iron removal, and incomplete mineralization [15].

And so, this requires the necessity of introducing a further better treatment step. Adsorption processes are commonly used as a final step in the treatment of industrial wastewater. Activated carbon (AC) has been proven as an efficient adsorbent for the removal of a wide variety of environmental pollutants. Because it is inexpensive and readily available. It can control color from the textile wastewater in an efficient manner [16]. Since adsorbent filter is an effective "treatment" process rather than traditional sand filter after Fenton process. The coupling of Fenton process with this adsorbent filter can be an appropriate alternative to achieve high levels of wastewater decontamination. That's why, the filtration unit prepared from the different layers of fine & coarse sand, gravel, activated carbon, charcoal will be able to adsorb more dissolved

organic solid from wastewater as they have more surface area to adsorb.

Current research work had performed to evaluate the removal efficiency of Adsorption filtration, Fenton process sequencing with sand filtration and adsorption filtration with activated charcoal for textile wastewater treatment. Before and after the treatment processes the physicochemical parameters pH, Dissolve Oxygen (DO), Electric conductivity (EC), Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD), Dissolve Organic Carbon (DOC) and Total Dissolve Solid (TDS) of the textile effluent was analyzed. In this study, whole treatment process was divided into three experiment. First experiment was conducted using adsorbent filtration by different layer of activated carbon, fine sand, coarse sand, and small stone. Then second experiment, Fenton process was applied with sand filtration. Third experiment was the application of Fenton process with adsorbent filtration process. Among this three-treatment process third experiment have shown better result than first and second one.

2. MATERIALS AND METHODS

2.1 Sample Collection

The raw effluent sample was collected from the Pakiza Dyeing and Printing Industries Limited. This is located at Savar Upazilla in Dhaka, Bangladesh. Water were preserved in the refrigerator at 4° C temperature for further physico-chemical analysis.

2.2 Preparation of Adsorbent Filter

Adsorbent filter was prepared by different layer of activated carbon, charcoal, fine sand, coarse sand, and small stone in a plastic jar. Activated carbon used for this experiment was purchased Osmosia Water from the Technology, Bangladesh. The layers were separated by net of very small size pores. Each layer is one centimeter (1 cm) thick. 5-6 small hole was made in the lower part of the plastic jar. Before making the layers the charcoal, sand and stones were washed very finely. After making the filtration unit, distilled water was passed through this filter several times for washing.

2.3 Physico-chemical Parameter Analysis

Color of the raw effluent was observed by naked eye and absorbance is measured by colorimetric method using UV-visible Spectrometer [17]. Decolorization efficiency of the treated sample was calculated using Equation (1) [11]:

Decolorization % = $(1-Abs_f/Abs_0) \times 100.$ (1)

where, Abs_f is the UV-absorbance of the treated dye and Abs_0 is the UV absorbance of the untreated dye.

DOC content of the sample was measured in high temperature catalytic oxidation (HTCO) method using TOC Analyzer (TOC-L CPN, Shimadzu, Japan). To measure DOC of four samples four vials were burned in muffle furnace for 120 minutes in 400° C. The samples were filtered in GF filter paper. An automated process injected the sample on a platinum catalyst at 680°C in an oxygen rich atmosphere. DOC removal efficiency was measured by equation (2) [11]:

Removal of DOC % = $(1-DOC_f/DOC_0)$. (2)

where, DOC_f is the Dissolve Organic Carbon of the treated dye and DOC_0 is the Dissolve Organic Carbon the untreated dye.

BOD ((mg/L) was determined by 5-days incubation (20 °C) method. The BOD values were found in automatically in computer. BOD removal efficiency was measured by equation (3) [11]:

Removal of BOD % = $(1-BOD_f/BOD_o)$ (3)

where, BOD_f is the chemical oxygen demand of the treated dye and BOD_0 is the chemical oxygen demand of the untreated dye.

COD (mg/L) was determined by gravimetric method and titrimetric method [17]. COD removal efficiency was measured by equation (4) [11]:

Removal of COD % = $(1-COD_f/COD_o)$ (4)

where, COD_f is the chemical oxygen demand of the treated dye and COD_0 is the chemical oxygen demand of the untreated dye.

Other physico-chemical parameter such as, EC and Salinity of wastewater was measured by conductivity meter. (HANNA Instrument, HI-8033), Turbidity was measured by turbidity meter (Microprocessor Turbidity Meter, HANNA Instrument: HI93703). The results were expressed in term of Formazin turbidity units (FTU). The pH of water was measured by using pH meter, Ecoscan Ion Meter (Model No.6). TDS was determined by using TDS Meter (HANNA.HI 8734 instrument). DO content of effluent samples were determined by using DO meter.

2.4 Procedure of Experiment

2.4.1 Experiment 1; Adsorption Filtration (AF)

In experiment-1, wastewater of textile industry was passed through the prepared adsorbent filter (activated carbon) and different parameters of treated water were measured. Adsorbent filter was prepared by making different layer of activated carbon, fine sand, coarse sand, and small stone in a plastic jar. The layers were separated by net of very small size pores. Each layer is one centimeter (1 cm) thick. 5-6 small pores were made in the lower part of the plastic jar. Before making the layers the charcoal, sand and stones were washed very finely. After making the filter distilled water was passed through this filter several times.

2.4.2 Experiment 2; Fenton Process + Sand Filtration (FP+SF)

In experiment-2, wastewater of textile industry was first treated by Fenton process and after that it was filtered by normal sand filter. For Fenton treatment, 500 mL samples were taken in a jar firstly then added iron catalyst (FeSO₄), 10 mL of FeSO₄ in ppm and then slowly added 3 mL of 30% concentrated hydrogen peroxide (conc. H_2O_2). Mixed the chemicals 5 mins and then settling time 30 mins kept for settlement [18]. Then the sample of wastewater filtered through normal sand filter (30 mins) and different physico-chemical parameters are measured. The chemicals used for the Fnton process were analytical grade and purchased from the HACH company.

2.4.3 Experiment 3; Fenton Process+ Adsorption Filtration (FP+AF)

In experiment-3, wastewater of textile industry was first treated by Fenton process and after that it was filtered by new adsorbent filter (activated carbon filter). For Fenton treatment, 10 mL of FeSO₄ was added in 500 mL wastewater samples in a jar then 3 mL 30% conc. H_2O_2 was slowly added. Mixed the chemicals 5 mins and then settling time 30 mins kept for settlement [18]. Then the sample wastewater filtered through new adsorbent filter (activated carbon filter) (30 mins) and different physico-chemical parameters were measured. The chemicals used

for the Fenton process were analytical grade and purchased from the HACH company.

3. RESULTS AND DISCUSSION

3.1 Characterization of Textile Wastewater

Effluent sample were analyzed for different physico-chemical parameters. The raw effluent was collected from the inlet of the textile dyeing and printing industry was dark green color and odor was pungent. Among the physico-chemical parameters of the textile effluent (Table:1), temperature of the wastewater sample was 31.5° C. EC value of raw sample was 2250 (µs/cm) which is almost two times greater than the recommended value of Bangladesh standard. Industrial effluent contains high ionic concentrations which is ultimately harmful for aquatic ecosystem [19]. TDS of raw sample was 870 mg/L which is within the range of standard value. Turbidity value of the sample was very higher than the recommended standard value and was 47.6 FTU. pH of the raw sample was basic in nature and the value was 10.14. the pH value of the raw effluent was not in the range of recommended standard limits (6.5 - 9). The salinity value of the raw of the raw sample 1200 mg/L. Higher salinity value of the wastewater is unfit for using in irrigation and industrial purpose. The DO value of the raw effluent was 1.8 mg/L which is below the limit of the recommended value. The lower DO content could be due to intrusion of high organic load in the water which leads to oxygen depletion [20]. The BOD and COD value of the raw sample was also very high indicating high degree of pollution. BOD of the sample was 250 mg/L, which exceeding the permissible standard limits. High BOD reflects high concentration of substances that can be biologically degradable. High level of BOD indicates the oxygen deficiency in water body causes serios problem to aquatic life. COD is the measure of oxygen equivalent of mater content of a sample that is susceptible to oxidation by strong chemical oxidant [21]. The COD value of raw effluent was 450 mg/L, which is almost two times greater than the recommended value. The DOC value of the raw effluent was 415 mg/L which also exceeds the tolerant level. DOC is a direct problem in drinking water because it affects color, odor, and taste of water, and can cause bacterial regrowth in the distribution system [22]. Moreover, the reaction of DOC with the most used disinfectant, chlorine, causes the formation of harmful disinfection by-product (DBPs) like trihalomethanes (THMs) [23]. These compounds are classified as probable or possible human carcinogens [24].

3.2 Effects of Different Treatment Process on Color Removal

At first the color of treated water was observed by naked eye and then UV absorbance was measured within 200nm-650nm wavelength. Finally, the color of treated water was become transparent after Fenton and adsorbent filtration process. UV visible spectrum also shows the less adsorption spectrum after treatment finally by Fenton process combined with adsorption filtration technique. From the Fig. 1, absorbance was reduced which means that the color of

wastewater was removed after treatment. In Fig. 1(a) absorbance was high which indicate the sample was highly colored and in Fig. 1(b) the absorbance was reduced that means color was removed after using only adsorption filtration. Then the sample was treated by Fenton process and normal sand filter and color was removed effectively has illustrated in Fig. 1(c). After that sample wastewater was treated by Fenton process and filtered by adsorbent filter and the absorbance was reduced thus color removal efficiency was highest among the treatment processes. From Fig. 1(d) it can be cleared that among the treatment process, process with adsorbent Fenton filtration (activated carbon filter) was more efficient for color removal.

Table 1. Physicochemical parameters of textile effluent





3.3 Changes of Parameters and Removal Efficiency

Changes of the Physicochemical parameters like, COD, DOC, Total Dissolve Solid (TDS), Dissolve Oxygen (DO), Electric conductivity (EC), and pH was measured before and after the treatment processes. Table 2 show the changes of parameters and removal efficiency after different treatment processes.

The DO level of raw effluent was too much low, which was 1.8 mg/L. After treatment, the DO level was gradually increased and the value became 5.0, 6.19, 6.61 after only filtration process, Fenton and normal sand filtration process, Fenton, and adsorbent filtration process, respectively which was within Effluent quality standard of Bangladesh [6]. The raw effluent collected from the inlet of the textile was alkaline and the value was 10.14. the pH value of the treated sample was decreased at 6.62, 4.5, 6.6 after only adsorption filtration process, Fenton and normal sand filtration process, Fenton. and adsorbent filtration process. respectively.

The Total Dissolved Solids (TDS) of raw effluent collected from the Textile Industry was 870 mg/L. TDS level was decreased into 280 mg/L, 220 mg/L, 150 mg/L after only filtration process, Fenton and normal sand filtration process, and adsorbent filtration Fenton. process, respectively. After Fenton process suspended solids greatly reduced, this due to the coagulant effect of Ferric iron (Fe³⁺) which is a good coagulant helpful in removing suspended solids after precipitation [3]. Electrical Conductivity (EC) of the raw effluent collected from the textile was high as 2250µs/cm. This value was decreased into 1560 µs/cm, 1010 µs/cm, 870 µs/cm respectively after only filtration process, Fenton and normal sand filtration process, Fenton, and adsorbent filtration process.

The BOD value of raw water was 250 mg/L which exceeds the recommended value of standard level. This value decreased into 180 mg/L, 70 mg/L, 50 mg/L after only filtration process, Fenton and normal sand filtration process, Fenton, and adsorbent filtration process, respectively. Experiment 3 have shown noteworthy result 92% removal efficiency for BOD.

The COD value of raw effluents of Pakiza Dyeing and Printing Textile Industry was 550 mg/L. This value was decreased into 300 mg/L, 180 mg/L, 110 mg/L after only filtration process, Fenton, and normal sand filtration process, Fenton, and adsorbent filtration process, respectively.

DOC value of raw wastewater was 415 mg/L. This value was decreased into 283 mg/L, 10.93 mg/L, 10.08 mg/L after only filtration process, Fenton, and normal sand filtration process, Fenton, and adsorbent filtration process. respectively. Experiment 2 and 3 both showed effectiveness for DOC removal. Maximum 97.57 % DOC removal efficiency was found after using Fenton process with adsorbent filtration. It has been found that after treatment by filtration process, Fenton, and normal sand filtration process, Fenton, and adsorbent filtration process pH, TDS, EC, BOD, COD, and DOC was come down gradually within Effluent quality standard of Bangladesh [6].

3.4 Comparative Analysis of Removal Efficiency of BOD and COD after Treatment Processes:

In Experiment-2, the sample wastewater was treated by Fenton process and filtered by normal sand filter and in Experiment-3, the sample wastewater was treated by Fenton process and filtered by adsorbent filter. From the Fig. 2 it has been seen that COD was decreased drastically after using experiment-3 (FP+AF) rather than Experient 1 and 2. Maximum 80% COD removal efficiency was found after using Fenton process with adsorbent filtration. That means the dye molecules and chromophore groups have destructed by free radical reactions produced from Fenton's [10]. After Fenton process, using activated carbon adsorption filtration has shown effective result. That means activated carbon has great adsorption capacity for the COD removal [25].

Fig. 3 shows that Maximum BOD removal efficiency was found from experiment-3 rather than Experiment-1 and 2. BOD removal efficiency is 80% in experiment-3 whereas 72% for experiment-2. That means this new adsorbent filter's performance is 8% more than normal sand filter. This adsorption filter was made by coarse sand, fine sand, charcoal and activated carbon. As charcoal and activated carbon has more surface area than sand and so, it makes better result during the adsorption in experiment-3.

Fig. 4 shows that Maximum DOC removal efficiency was found from experiment-2 and 3 rather than Experiment-197.36% and 97.57% respectively.

Parameters	Before Treatment Raw effluent	After Treatment						
		Experiment- 1(AF)	Removal Efficiency (%)	Experiment- 2(FP+SF)	Removal Efficiency (%)	Experiment- 3(FP+AF)	Removal Efficiency (%)	
рН	10.14	6.62	-	4.5	-	6.6	-	
DO (mg/L)	1.8	5	-	6.19	-	6.61	-	
TDS (mg/L)	870	280	67.81	220	74.71	150	82.75	
EC (µs/cm)	2250	1560	30.66	1010	55.11	870	61.33	
BOD (mg/L)	250	180	28	70	72	50	92	
COD (mg/L)	550	300	45.45	180	67.27	110	80	
DOC (mg/L)	415	283	31.80	10.93	97.36	10.08	97.57	

Table 2. Changes of parameters and removal efficiency after different treatment processes



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Fig. 2. COD Removal efficiency after experiment 1, 2 and 3

EXPERIMENT 1, 2 & 3

I	Fig. 3. BOD Removal efficie	ncy after experime	ent 1, 2 and 3
Table	3. Comparative removal effi	iciency with other	treatment process

Treatment process	Color removal	COD removal	Reference	
Fenton oxidation with membrane separation	99%	75%	[11]	
Fenton Reaction	84.66%	78.14	[3]	
Fenton-coagulation	99%	80%	[26]	
Fenton-Coagulation Flocculation	-	74%	[9]	
Adsorption- Fenton Oxidation	-	93%	[25]	
Experiment 3(FP+AF)	78.57%	80%	This study	



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Fig. 4. DOC Removal efficiency after experiment 1, 2 and 3

3.5 Removal Efficiency of Other Treatment Process

From this study it was found that the Fenton process with adsorption filtration has good textile wastewater treatment capacity than other treatment process (Table. 3). literature review shows that the Advance treatment process like Fenton Process is very much effective for COD and color removal. Researcher applied Fenton process with Membrane separation, Coagulation-Flocculation and Adsorption. Among them Adoption-Fenton Oxidation was more effective for COD (93%) removal [25]. Present study also found more effective result for color and COD (80%) removal using Fenton process with Adsorption filtration. Maximum color removal efficiency after treatment (Experiment-3) was 78.57% which shows a good color removal from textile effluent. This result is very close to the result 84.66 % found by [3]. So, Fenton process can be applied in the industrial scale as an environment friendly, economic, and effective alternative.

4. CONCLUSION

The textile dyeing wastewater is one of the most important sources of environmental pollution. From the physicochemical analysis it has assumed that this wastewater has the characteristics of high level of color, BOD (250 mg/L), COD (550 mg/L), EC (2250 µs/cm) TDS (870 mg/L), DOC (415 mg/L), and deficiency of Dissolve oxygen (1.8 mg/L). If being directly discharged without being treated, it will bring serious threat to the ecological environment. From this experiment impressive removal efficiency of different parameters such as Color, EC, TDS, pH, DO, BOD, COD, DOC, from the untreated textile effluents was observed which was within recommended level of Bangladesh. Fenton process combined with adsorbent filter has shown more efficiency for removing color (78.57%), EC (61.33%), TDS (82.75%), BOD (92%), COD (80%), and DOC (97.57%) than normal sand filter.

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DISCLAIMER

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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